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New Data for Aerosols Generated by Releases of Pressurized Powders and Solutions in Static Air

ORIGINAL

Prepared by M. Y. Ballinger, S. L. Sutter, W. H. Hodgson

Pacific Northwest Laboratory
Operated by
Battelle Memorial Institute

Prepared for
U.S. Nuclear Regulatory Commission

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New Data for Aerosols Generated by Releases of Pressurized Powders and Solutions in Static Air

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EXECUTIVE SUMMARY

Pacific Northwest Laboratory conducted three sets of experiments to measure aerosols generated by pressurized releases. These aerosols are generated from releases of uranine pressurized with carbon dioxide, flashing sprays, and low-pressure depleted uranium dioxide powder releases. The purpose of these experiments was to supplement data from earlier experiments investigating the source term from pressurized releases. These experiments are part of a program to develop source-term estimation methods for analysis of accidents in nuclear fuel cycle facilities.

Mass airborne and particle size distribution of aerosols generated by CO_2/uranine releases, flashing sprays of uranine, and low-pressure depleted uranium powder releases were measured. Source volume and release pressure varied in each set of experiments. Source volumes were 100 and 350 mL for CO_2/uranine releases and flashing sprays and 100 and 350 g for low-pressure powder releases. Release pressure varied from 9 to 500 psig.

The mass airborne from CO_2-pressurized releases was larger than those from comparable air-pressurized systems. However, the differences in mass airborne from CO_2- and air-pressurized systems was not as great as anticipated, based on the amount of gas dissolved in the liquid and the volume of liquid ejected from the pressurized-airborne release equipment (PARE) for both systems.

Flashing sprays produced a much larger source term than all other pressurized releases performed under this program. All but one of the flashing-spray releases resulted in all of the liquid being evaporated or ejected from the PARE.

Low-pressure powder releases provided data in the energy region between previously reported spills and pressurized releases. Mass airborne increased with solution volume and release pressure as expected.

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1.0 INTRODUCTION

Aerosol generation characteristics of accidental particle releases in nuclear fuel cycle facilities are being investigated at Pacific Northwest Laboratory (PNL). The work is sponsored by the U.S. Nuclear Regulatory Commission, Office of Nuclear Materials Safety and Safeguards. Safety analysis reports and environmental impact statements must evaluate the consequence of postulated accidents in or involving the facility in question. During an accidental release from a nuclear fuel cycle facility, the dominant pathway to man is usually through airborne particles, so it is necessary to determine an aerosol source term (i.e., the quantity of material initially airborne from an accident).

Earlier work (Sutter et al. 1981; Sutter 1983) reported on free-fall spills and pressurized releases of powders and liquids in static air. This report covers results of additional pressurized-release experiments conducted as part of an ongoing study of accident-generated aerosols. These experiments dealt with pressurized releases of liquid using a soluble gas, flashing sprays, and pressurized releases in the energy region between previously reported spills and pressurized releases.

Sutter reported that liquid releases from the pressurized-airborne release equipment (PARE), using air as a pressurizing gas, were lower than anticipated. High-speed photography revealed that the release seemed to be a possible atomization of the liquid caused by the rapid depressurization. The only noticeable release was a rapidly moving cloud, and almost all the solution volumes remained in the PARE. These findings lead to speculation that the mass airborne may be a function of the amount of gas dissolved in solutions.

To test this theory, carbon dioxide, a highly soluble gas, was used to pressurize contained volumes of sodium fluorescein (uranine) solutions. The gas was injected through openings in a ring below the liquid surface to ensure mixing. Two solution volumes were released at three pressures: 50, 250, and 500 psig. These are the same parameters used in earlier experimental work, thus, allowing us to compare results. Airborne samples were collected for a half-hour following the event with high-volume impactors and a cascade impactor.

Flashing sprays provided data on the source term from rapid releases of hot liquids. This kind of release could result in a much higher source term than other pressurized releases. Three uranine solution volumes of 100, 350, and 700 cm^3 were heated using a heat tape on the outside of the PARE. Releases occurred when the vapor pressure of the liquid reached 57, 124, and 240 psig. Airborne samples were collected with CO_2-pressurized releases.

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Previous experiments provided source term data in the low-energy region (spills where energy is caused by gravity) and in the high-energy region (pressurized releases at 50 psig or greater where the energy is caused by elevating the gas pressure). Low-pressure releases of depleted uranium dioxide (DUO) powder were performed to investigate releases in the energy region between the two extremes. Source quantities of 100 and 350 g DUO were used for easier comparison with previous work. Releases were made at 9, 17.5, and 24.5 psig, and airborne samples were collected as with CO_2-pressurized releases and flashing sprays.

Experimental procedures are detailed in Chapter 2, followed by a discussion of the results in Chapter 3. Earlier work (Sutter et al. 1981) described pressurized release experiments performed at PNL. The same release vessel, source materials, pressure chamber (with modifications for flashing sprays and CO_2 pressurized releases), sampling procedures, and sample analysis techniques used in these previous experiments were used here.

Airborne mass measurements and median aerodynamic equivalent diameters (AED) are listed in Appendix A. Calculation of solubilities of gases in uranine solutions supporting the experiments using CO_2 releases are given in Appendix B. Data from these experiments will be used in ongoing work to develop release models based on physical parameters.

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2.0 EXPERIMENTAL PROCEDURES

Experiments were performed to determine the amount of airborne material and the aerodynamic particle size of the aerosols generated by three types of releases: 1) pressurized release of a liquid from a chamber pressurized with carbon dioxide gas, 2) flashing sprays of superheated liquids, and 3) low-pressure release of powdered DUO. These releases took place in static air. Total release from the event was measured; the time-dependent behavior of the aerosol was not investigated. The aerosols were contained in a stainless steel vessel approximately 3-m high and 2.9 m in dia with a 20-m^3 volume, about the size of a small room. This vessel is the radioactive aerosol release tank (RART).

2.1 CO_2/URANINE RELEASES

The PARE used in earlier work was modified for use with CO_2, as shown in Figure 2.1. This modification made it possible to fill the entire chamber with

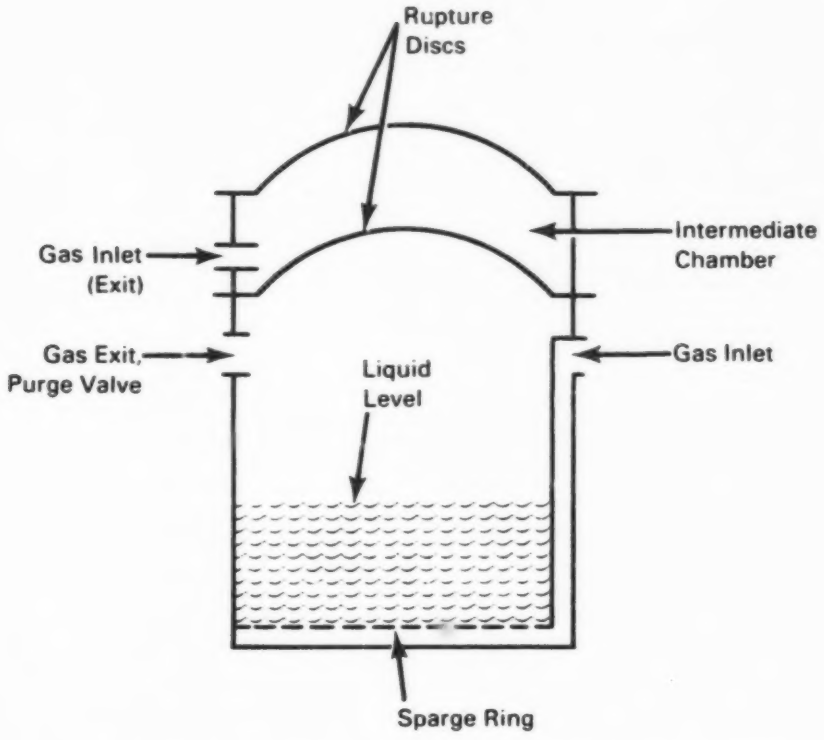

FIGURE 2.1. PARE Modification for CO_2/Uranine Release Experiments

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the pressurizing gas and saturate the contained liquid before the release. The gas enters the pressure chamber through 1/4-in. stainless steel tubing formed into a ring at the bottom of the chamber. To allow intimate mixing of the entering gas and the contained liquid, 42 openings, 1/16 in. dia, were drilled in the ring. The lower chamber was filled with CO_2 gas, and air was displaced before the liquid was ejected. This chamber was equipped with a purge valve to accomplish the air displacement. After eliminating air from the PARE, the purge valve was closed and the chambers incrementally pressurized to the preselected experimental level at which the disks were ruptured.

Carbon dioxide was selected as the pressurizing gas because it is highly soluble in aqueous solutions. Abc . 8 to 10 chamber volumes of the gas were run through the PARE before pressurization. The mole fraction of CO_2 dissolved in the uranine solution was calculated using Henry's Law with corrections for pressure. The calculations are shown in Appendix B. Values for air were also calculated to enable comparisons with earlier work.

Results of the calculations for both the air- and CO_2-pressurizing systems are listed in Table 2.1 and show that 50 times more CO_2 than air is potentially dissolved in the solutions. Deviations from the calculated values can result from chemical reactions of the gas with the solutes in the liquid. The CO_2 solubility could be affected in an unknown way by the nonvolatile solute uranine.

TABLE 2.1. Estimated Mole Fraction of Test Gases Dissolved in Uranine Solution[a]

Pressure, psig	Test Gas	Mole Fraction Dissolved
500	Air	4.2×10^{-4}[b]
	CO_2	2.0×10^{-2}
250	Air	2.3×10^{-4}
	CO_2	1.3×10^{-2}
50	Air	4.8×10^{-5}
	CO_2	2.5×10^{-3}

(a) Assume solubility of gases the same as in water.
(b) Air value calculated assuming 78% N_2, 21% O_2; inert remainder not included.

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2.1.1 Experimental Matrix

Uranine solutions with a concentration of 10 g/L were used for these experiments. Two volumes (100 and 350 cc) and three pressures (50, 250, and 500 psig) were used to enable direct comparison of the results with earlier work (Sutter 1983). Experimental pressures were monitored with a transducer indicator that was calibrated before use to an accuracy of plus or minus 0.5%. The chamber pressure was displayed as a digital readout.

2.1.2 Procedure

The general sampling and experimental procedure used in conducting the experiments is shown in Figure 2.2. First, both chambers were pressurized, then the rupture discs were broken by depressurizing the intermediate chamber, and the liquid was ejected. Samplers were immediately turned on and the particle-laden air pulled onto filters, as indicated by the solid arrows in Figure 2.2. Clean air entered from the rear, as indicated by the light arrows. These experiments were conducted in the RART.

High-volume samplers equipped with glass fiber filters were used for mass sampling; a cascade impactor was used for size distribution samples. These samplers were located at the same levels as in previous experiments [i.e., high-volume samplers were at the 1- and 2-m level, and the cascade impactor was

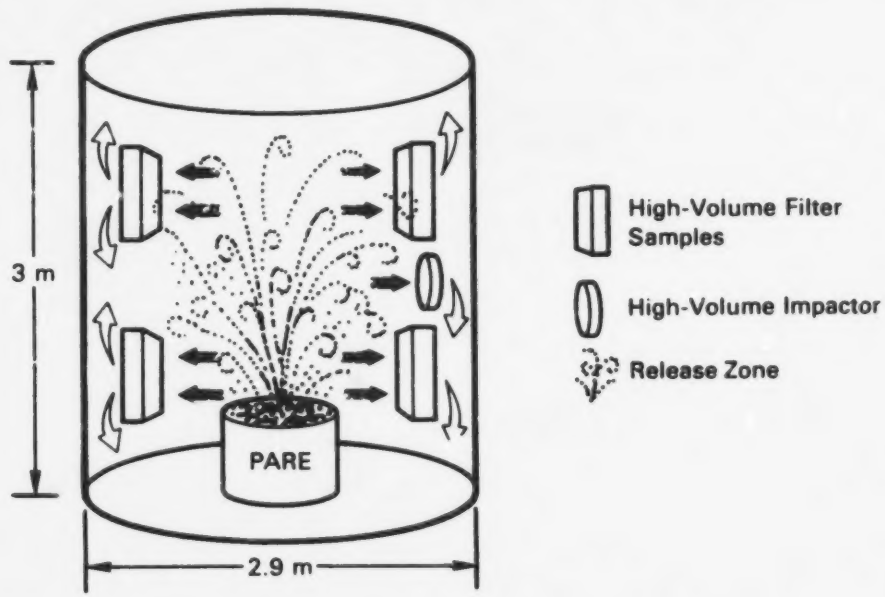

FIGURE 2.2. Sampling for a Pressurized Release with High-Volume Samplers

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at the 1.5-m level (Figure 2.2)]. Eight RART volumes of air were pulled through the samplers in a 30-min run; this is the same sampling time used for earlier liquid releases.

High-speed photography was also used to better understand results. Visual records can confirm and help explain the data analysis results.

2.1.3 Sample Analysis

Uranine collected on the filters was dissolved in water and analyzed by use of a calibrated fluorometer. The lower detection limit is about 1×10^{-9} g/cm^3. Equations for the measured values at each of four fluorometer ranges were formulated and used for calculating results. The R^2 values (correlation between observed and predicted values) for each attenuation curve approached 1, indicating a good fit for the calibration standards.

Carbon dioxide used as a pressurizing gas could acidify the sample solutions and lead to problems with the analysis. Fluorescein fluorescence intensity increases sharply above a pH of 3 and is fairly constant between pH 6 and 8 (Feverstein and Selleck 1963). Therefore, the pH of each collected sample was measured before the sample was analyzed to ensure that it was in the neutral range. All solutions were neutral at the time they were collected. The results of the analysis is discussed in Section 3.1.1.

2.2 FLASHING-SPRAY RELEASES

The PARE used in earlier work was modified, as shown in Figure 2.3, to permit measurement of temperature of the liquid and the vapor pressure above the liquid surface. A 1/16-in.-dia sheathed thermocouple was inserted through the bottom air inlet and sealed with a 1/8-in.-dia tubing fitted with a Teflon® compression gasket. Use of the Teflon® gasket permitted adjustment of the thermocouple location.

A pressure transducer was located about 16 in. from the PARE and was attached to the PARE through a 1/4-in.-dia steel tubing with a "T" to permit pressure relief into the atmosphere with a remotely operated solenoid valve. An extension of this tube placed the opening above the liquid surface level when 700 mL of uranine was placed in the PARE.

Two 4-ft sections of 1-in.-wide heating tape were wrapped around the PARE body. The heating tape and other exposed metal surfaces were surrounded by loosely packed glass wool. Electrical energy for the heat tapes was remotely controlled with variable transformers.

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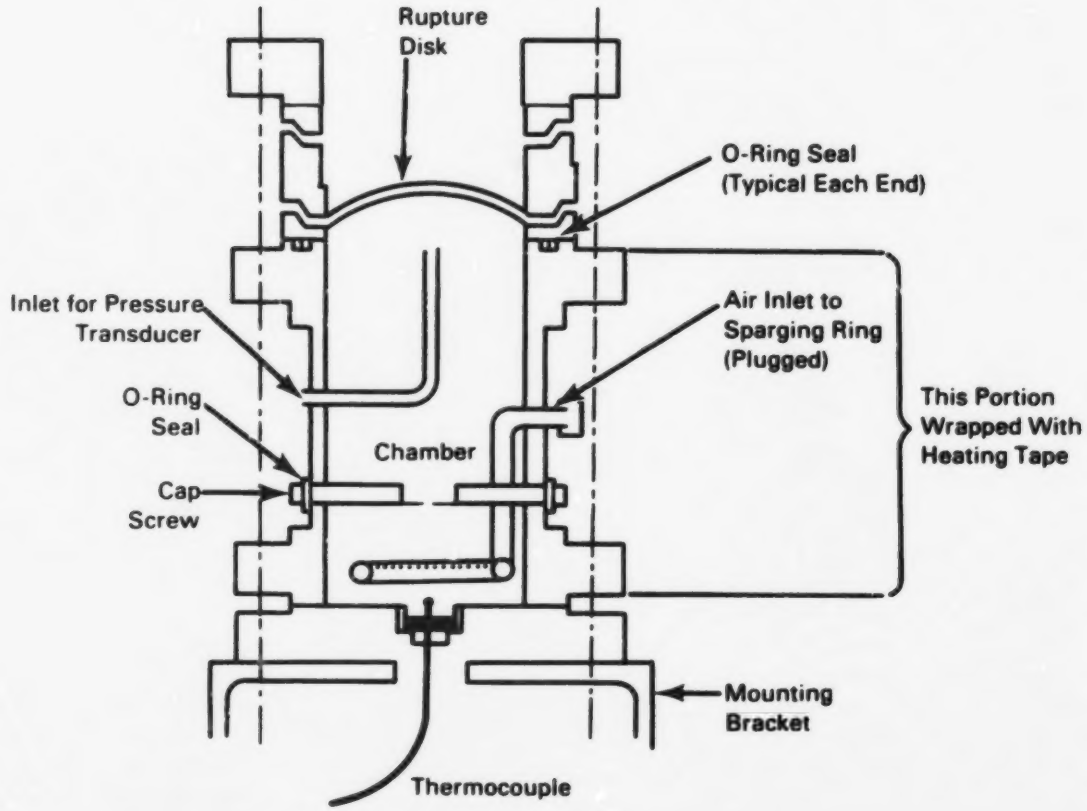

FIGURE 2.3. PARE Modifications for Flashing-Spray Release Experiments

Uranine solutions with a concentration of 10 g/L were used for these experiments. Three volumes of 100, 350, and 700 cm^3 of liquid were used. The 100 and 350 cm^3 volumes enabled direct comparison of the results with previous experiments. Releases occurred at 50, 100, and 250 psig.

The rupture pressure for these experiments was the vapor pressure of the superheated liquid. A computer code was developed to predict temperatures that would result in rupture of the PARE rupture discs at pressures ranging near those used in previous releases. (The code calculated vapor pressure from given solution volume and temperature.) A plot (Figure 2.4) of the pressure in pounds per square inch gage versus the temperature in degrees Celsius was constructed from the data. The temperature required to attain rupture near the desired pressure was read from the plot. Also shown in Figure 2.4 are the temperatures measured during the actual runs. Except for run 5, measured temperatures came very close to those predicted. In run 5, the thermocouple may have malfunctioned. Approximate liquid surface temperatures at rupture were 134, 161, and 202°C. The thermocouple used to monitor temperature was

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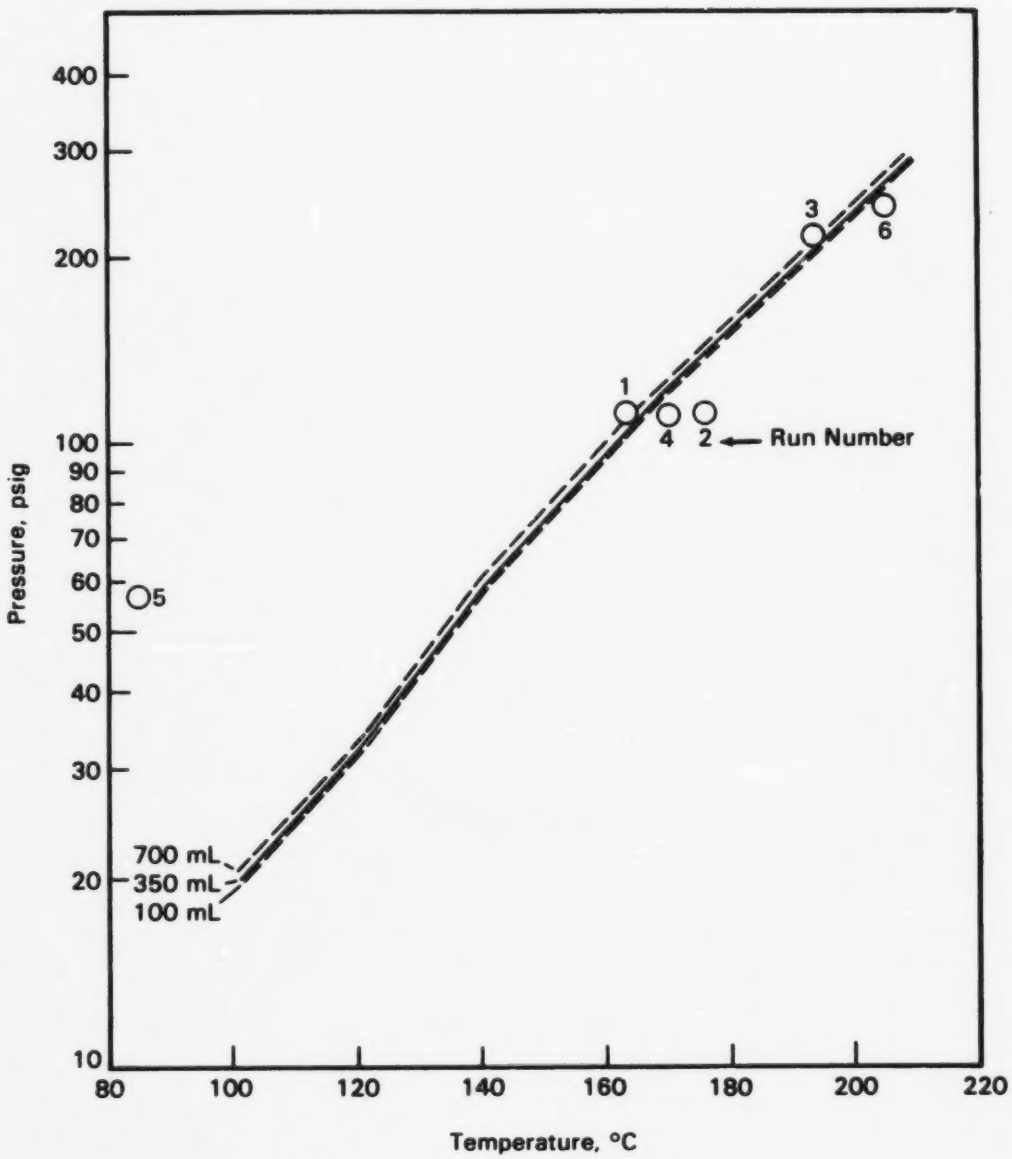

FIGURE 2.4. Measured and Predicted Temperatures for Flashing-Spray Releases

located approximately 1-1/4 in. below the liquid surface before heating. Temperature was read on a potentiometer located outside the RART.

Heat was supplied to the PARE at a relatively constant rate by setting the variable transformers at 85% of maximum. Heating continued until the disk ruptured. Immediately, the heat was turned off and the samplers turned on. Sampling and analysis was the same as that for the CO_2-pressurized releases.

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2.3 LOW-PRESSURE POWDER RELEASES

The pressurized airborne release equipment used in the CO_2/uranine and earlier releases was modified slightly for the low-pressure powder releases (Figure 2.5). The sparging ring that permitted gas to be bubbled through the uranine solution was sealed at the inlet connection. Depleted uranium dioxide powder was suspended on filter paper over a screen about 2 1/2 in. above the air inlet that was in the base of the PARE. A single rupture disk was located in the lower rupture disk flange.

Pressures of 9, 17.5, and 24.5 psig were selected to explore the energy region between the previously reported spill and pressurized release experiments. Experimental pressures were monitored with a transducer indicator, which was calibrated before use to an accuracy of ±0.5%. The chamber pressure level was displayed as a digital readout. To allow direct comparison with previous experiments, 100 and 350 g of DUO powder were used as source quantities.

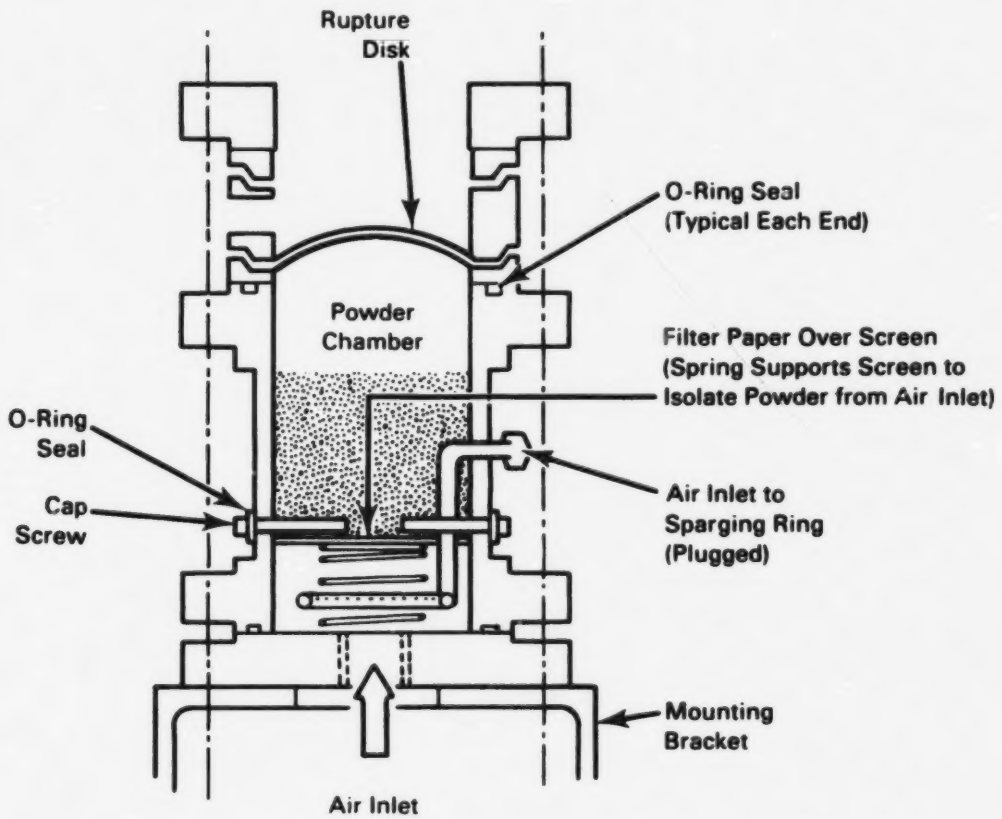
Rupture Disk
O-Ring Seal (Typical Each End)
Powder Chamber
Filter Paper Over Screen (Spring Supports Screen to Isolate Powder from Air Inlet)
O-Ring Seal
Cap Screw
Air Inlet to Sparging Ring (Plugged)
Mounting Bracket
Air Inlet

FIGURE 2.5. PARE Modification for Low-Pressure Powder Releases

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Compressed air was used as the pressurizing gas and release was accomplished by rapidly increasing pressure until rupture occurred. Sampling was similar to that for CO_2-pressurized releases.

Filter samples containing collected uranium aerosols were dissolved in acid and analyzed using laser fluorometry. This method employs a pulsed nitrogen laser to excite uranium in a solution containing a pyrophosphate reagent. Fluorescent signals are amplified and integrated, and the results are displayed on a meter. Previous experiments established the experimental error to be less than 10%.

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3.0 RESULTS

Included in this chapter are the results of the high-volume sampling of pressurized releases of liquid using CO_2 as the pressurizing gas, flashing sprays, and low-pressure releases of DUO. These results will be used to develop source-term estimation models based on controlling parameters. The models will be described in a later report.

3.1 CO_2/URANINE RELEASES

Complete listings of the measured values for the uranine mass airborne are tabulated in Table A.1 in Appendix A. Values for particle size are found in Table A.2.

3.1.1 Mass Airborne

The total uranine mass airborne is listed in Table 3.1. The table includes average releases from air-pressurized solutions. Carbon dioxide-pressurized releases were at the same or higher levels as air values and range up to eight times greater.

Carbon dioxide was used as a pressurizing gas to test the theory that the amount airborne is a function of the amount of gas dissolved in solution. Appendix B shows the method of calculating the quantity of CO_2 dissolved in uranine for the CO_2-pressurized releases. This Appendix also gives calculations for the air-pressurized releases and for flashing sprays. When the three

TABLE 3.1. Uranine Mass (Grams) Airborne from Air- and CO_2-Pressurized Releases

Chamber Pressure, psig	350 cm^3		100 cm^3	
	Air	CO_2	Air	CO_2
500	1.67×10^{-3}	4.7×10^{-3}	1.5×10^{-3}	2.2×10^{-3}
250	3.3×10^{-4}	1.4×10^{-3}	6.0×10^{-4}	5.0×10^{-3}
50	2.8×10^{-5}	4.9×10^{-5}	5.0×10^{-5}	5.5×10^{-5}

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sets of experiments are plotted (Figure 3.1), the above theory does not appear to hold true for CO_2-pressurized releases. Several factors may account for this:

- CO_2-pressurized releases may have produced large particles, which settled out more quickly than the air-pressurized particles, thus less of the initially airborne solution was measured by the impactors. The fact that much more liquid was initially ejected from the PARE with CO_2 releases than with air releases corroborates this theory.

- CO_2 may have lowered the pH of the uranine sufficiently to precipitate some of the tracer out of solution before release. If this occurred, the amount airborne measured by the impactors may be underestimated since it would be based on a more dilute solution than was

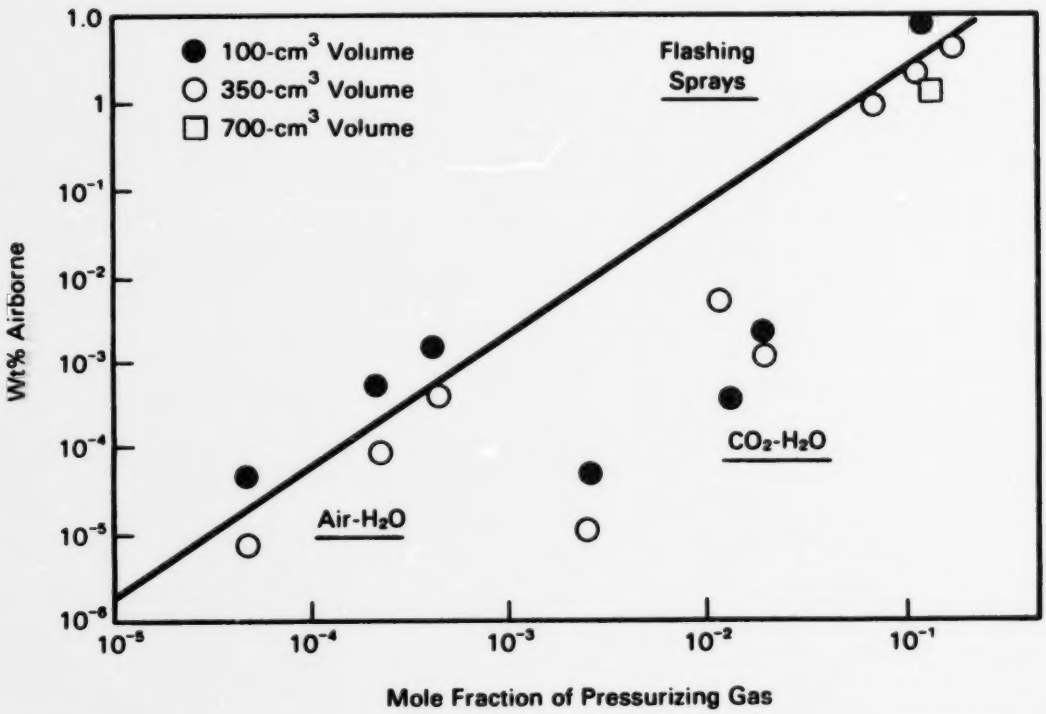

FIGURE 3.1. Weight Percent Airborne as a Function of Mole Fraction of Pressurizing Gas

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originally put in the PARE. The presence of an orange precipitate in the liquid left in the PARE during one of the runs substantiates this theory.

- The dissolved CO_2 is not released as quickly as the dissolved air because of a slow reverse hydrolysis of CO_2 and its reaction products with water. For example, the beverage industry depends on carbonated drinks retaining their effervescence for some time after opening.

- Much of the CO_2 must have stayed in solution for some time after release. Therefore, not all of the gas dissolved in the liquid contributed to the energy available to form aerosols immediately on release. The orange precipitate mentioned above was stable in a closed container but went back into solution when the container was left open to the air. The conclusion can be made that CO_2 caused sodium fluorescein to precipitate by dropping the pH of the solution to less than 4. As long as the container was closed, no CO_2 was allowed to escape, and the precipitate remained. When the container was open, CO_2 went out of solution, the pH increased, and the precipitate redissolved.

The actual airborne mass could have been higher than that measured at the upper pressures since the release impacted on the RART ceiling and spread to the task walls, dripping down the sides. In an unenclosed space, much of the solution on the ceiling would probably have become airborne. On the other hand, the enclosure effect could also have enhanced the release since the liquid column impacting on the ceiling could have generated airborne droplets.

3.1.2 High-Speed Photography and Liquid Ejected

High-speed photography revealed that this CO_2-pressurized release is a different type of event than the air-pressurized release as a geyser effect was evident. Liquid could be seen ejected from the PARE, unlike the cloud that was produced from the air-pressurized system and was barely noticeable.

The amounts ejected from the PARE are compared for air and CO_2 in Table 3.2 and illustrated in Figure 3.2. The percent of source ejected was as high as 85% for CO_2 (500 psig, 350 cm^3) compared to 4% for the corresponding air-pressurized system. In Figure 3.2, the volume ejected is indicated on the left ordinate; on the right is the percent ejected.

However, these measurements do not reveal the entire phenomena. The high-speed photography of CO_2 pressurized releases showed that the solution jetted from the chamber, reached an apex, and fell back, with a portion falling

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TABLE 3.2. Liquid Volume Ejected from the PARE

Pressure, psig	350 cm^3				100 cm^3			
	Air		CO_2		Air		CO_2	
	cm^3	% of Source	cm^3	% of Source	cm^3	% of Source	cm^3	% of Source
500	14	4	298	85	5	5	52	52
250	8	2	225	64	6	6	33	33
50	7	2	25	7	3	3	5	5

directly back into the chamber. Thus, some of the amount left in the PARE (all of which was assumed to have not been ejected) may have initially been airborne.

The liquid remaining within the PARE and that which pooled on the floor of the RART contained solid orange particles and was foamy. The solid particles may be precipitated fluorescence. Following the release, the foam dissipated, and the solid became redissolved.

3.1.3 Weight Percent Airborne

Release models under development at PNL estimate the weight percent of the source airborne. The weight percent airborne values generated in these experiments are listed in Table 3.3 along with the corresponding values for the air-pressurized system.

The weight percent airborne was usually larger for the smaller source for both air and CO_2. Percent airborne values ranged from 0.0008 (0.0028 g from 350 cm^3 of liquid, air pressure 50 psig) to 0.5% (0.5 g from 100 cm^3 liquid, CO_2 pressure 250 psig). The values increased with pressure as anticipated, with the exception of the 250 psig release from a 100 cm^3 source. However, the 500 psig release might have been higher in a larger enclosure that did not allow ceiling deposition or lower if significant numbers of small particles were produced by the impact.

3.1.4 Particle Size Airborne

Median AED particle sizes generated by the air- and CO_2-pressurized liquid releases are listed in Table 3.4 and are in the same range for both gases. However, the σg values for the CO_2-pressurized releases range from 2.6 to 11 compared to the range of average values of 1.9 to 3.5 for air. This range of

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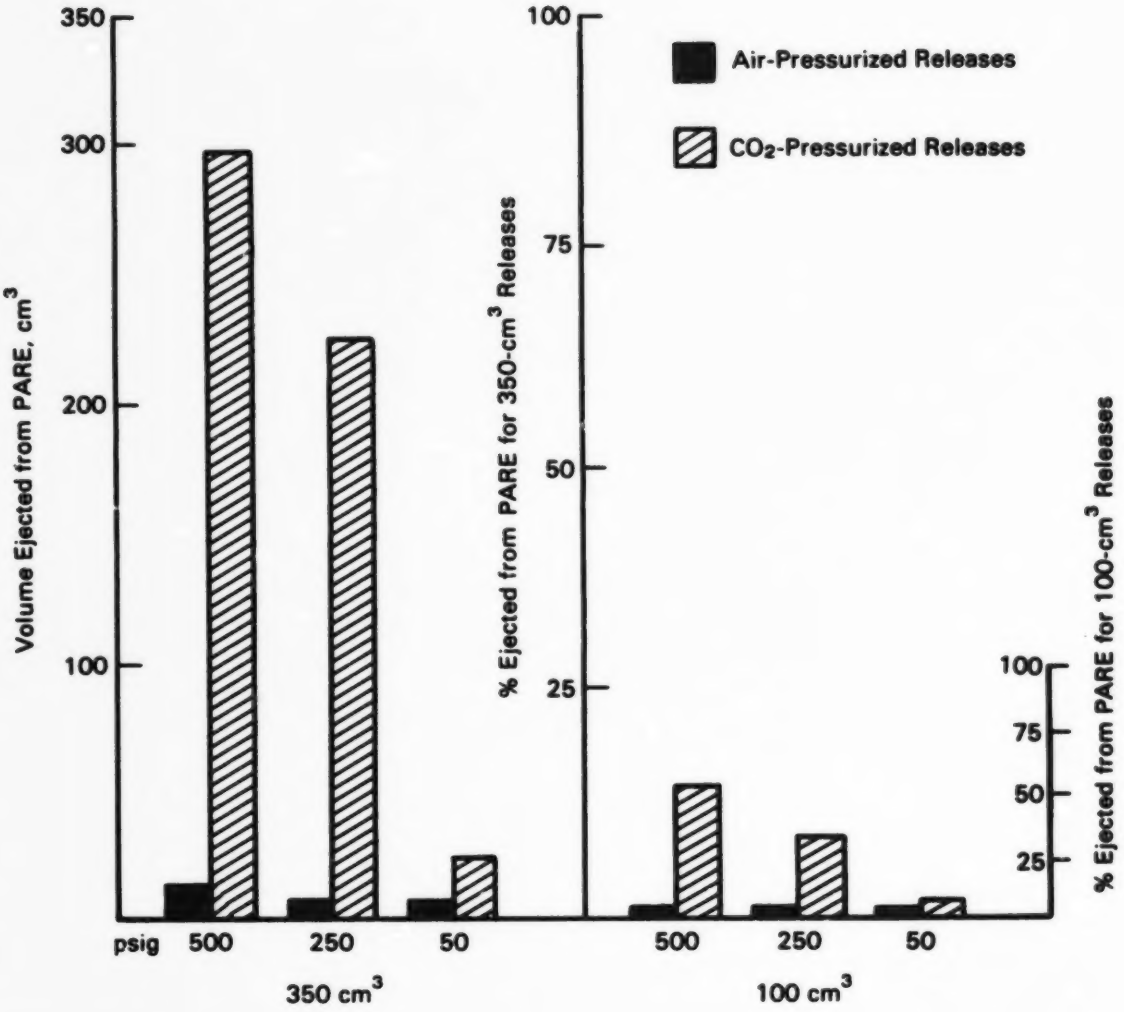

FIGURE 3.2. Solution Volumes Ejected from PARE in Air- and CO_2-Pressurized Releases

TABLE 3.3. Weight Percent Airborne from Air- and CO_2-Pressurized Releases

Chamber Pressure, psig	350 cm^3		100 cm^3	
	Air	CO_2	Air	CO_2
500	0.05	0.13	0.15	0.22
250	0.01	0.039	0.06	0.50
50	0.0008	0.014	0.005	0.006

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TABLE 3.4. Median AED of Particles Generated by Air- and CO_2-Pressurized Liquid Releases

Chamber Pressure, psig	350 cm^3				100 cm^3			
	Air		CO_2		Air		CO_2	
	μm	σg	μm	σg	μm	σg	μm	σg
500	4	2.7	3.3	11	4	3.5	2.7	5.5
250	4	2.3	2.3	5.2	3	3.8	2.1	2.6
50	2	3.5	1.5	3.8	2	4	1.9	3.3

values indicates that the air vaporization produced a more uniform aerosol; it could also indicate that there is more than one particle formation process operating in the CO_2 release.

High-speed photography verified the experimental results. The CO_2 produced a column of uranine solution, which broke off into large chunks and droplets. While a large fraction of the source was ejected, it did not produce a large amount of fine aerosol as might have been anticipated. Energy input is required to eject the liquid and produce fine particles.

The particle-size distributions were measured by using the cascade impactor samples and may not be representative of the size distribution of the initial airborne liquid droplets. Significant evaporation of the droplets before they are captured in the impactor could mean that the measured size distribution does not correspond to that of the actual airborne aerosol. Corrections to the size distribution will be handled in model development.

3.2 FLASHING-SPRAY RELEASES

Complete listings of the measured values for the uranine mass airborne of superheated liquids are tabulated in Table A.3 in Appendix A. Values for particle size are found in Table A.4 in Appendix A.

3.2.1 Mass Airborne

Total uranine mass airborne for the flashing-spray experiments is listed in Table 3.5. The mass of uranine made airborne in these experiments is about 100-fold greater than in previous pressurized gas releases. The additional energy provided by the elevated temperature was sufficient to eject the liquid and produce the fine particles that remained suspended.

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TABLE 3.5. Mass of Uranine Made Airborne by Flashing Sprays of Superheated Liquid

Chamber Pressure, psig	700-cm^3 Source, g	350-cm^3 Source, g	100-cm^3 Source, g
240		1.58×10^{-1}	
124	9.68×10^{-2}	7.12×10^{-2}	7.83×10^{-2}
57		2.85×10^{-2}	

3.2.2 Liquid Ejection

As with previous experiments involving pressurized-uranine solutions, some of the solution impacted on the RART ceiling and spread to the walls. Only one run (57 psig) had uranine left in the PARE after release. For this run, 76% of the 350-cm^3 source was ejected from the PARE. The residual heat in the PARE following the release may have evaporated remaining uranine solution from most of the runs; however, visual observation of the interior of the PARE following sampling did not show a large amount of residue, which would be expected if significant evaporation took place.

3.2.3 Weight Percent Airborne

The values for weight percent of uranine airborne in the liquid-pressurized release experiments are plotted in Figure 3.3. Weight percent uranine airborne from flashing sprays is listed in Table 3.6. Values for flashing sprays are one and one-half to two orders of magnitude higher than for air- and CO_2-pressurized releases.

The amount of pressurizing gas in the liquid was calculated for flashing sprays (see Appendix B). Figure 3.1 shows a plot of weight percent airborne versus mole fraction of pressurizing gas for three sets of data: flashing sprays, air-, and CO_2-pressurized releases. A straight line with correlation coefficient (R^2) of 98% can be drawn through the air-pressurized releases and flashing-spray data as shown in the figure. The source term from these two sets of experiments seems to be influenced by the amount of gas dissolved in the liquid. The measured amount of airborne from the CO_2-pressurized releases may be conservative for the reasons specified in Section 4.3.

3.2.4 Particle Size Airborne

Median AED particle sizes generated by flashing-spray releases are listed in Table 3.7 and are approximately double the values for air- and CO_2-pressurized releases.

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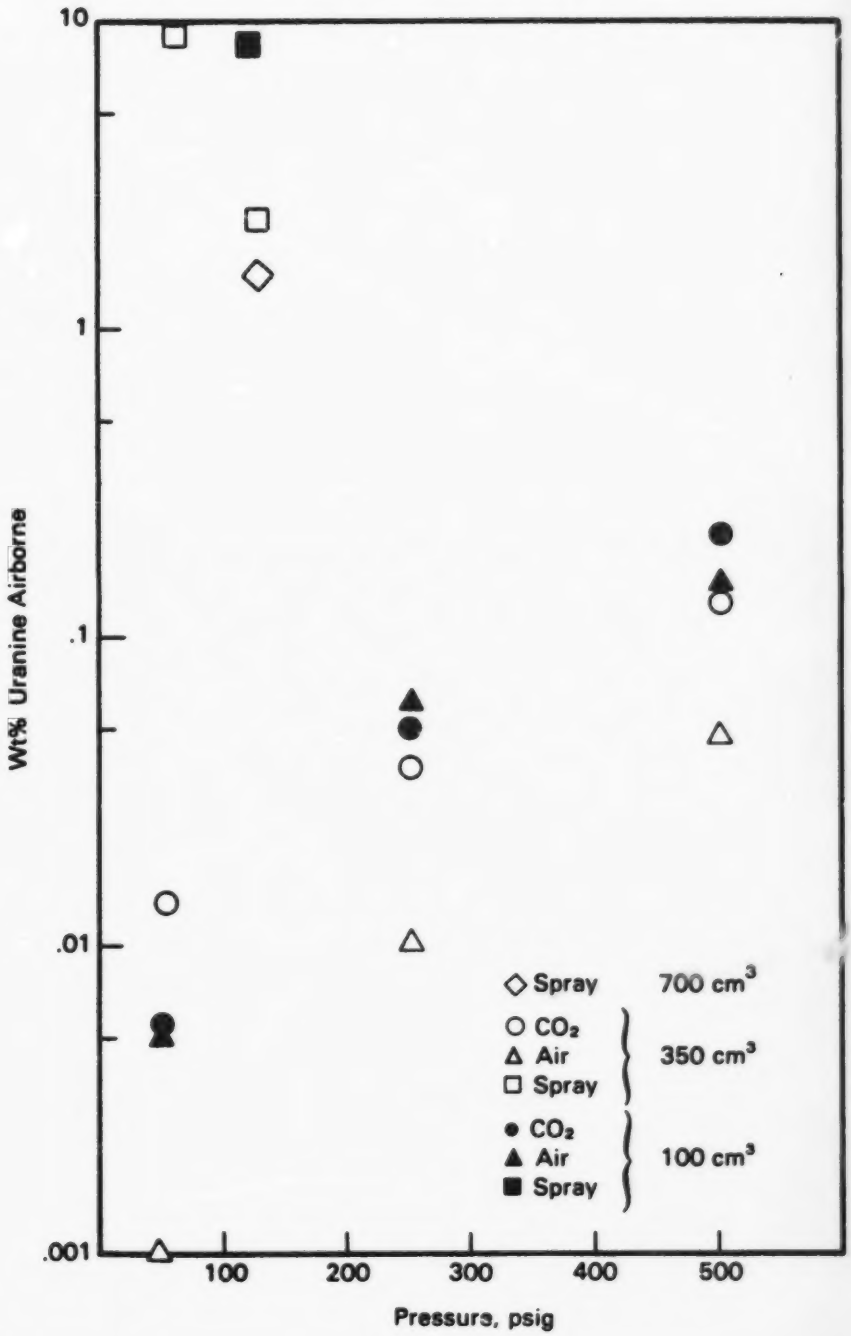

FIGURE 3.3. Weight Percent of Uranine Airborne in Liquid-Pressurized Releases

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TABLE 3.6. Weight Percent Uranine Airborne from Flashing-Spray Releases

Chamber Pressure, psig	700-cm^3 Source	350-cm^3 Source	100-cm^3 Source
240		4.96	
124	1.51	2.22	8.51
57		0.89	

TABLE 3.7. Median AED of Particles Generated by Flashing-Spray Releases

Chamber Pressure,	700-cm^3 Source		350-cm^3 Source		100-m^3 Source	
psig	AMMβ μm	σ_g	Ammβ μm	σ_g	Ammβ μm	σ_g
240			6.25	2.7		
124	5.9	2.6	7.7	2.7	6.4	3.3
57			8.4	5.6		

The intermediate- and high-pressure releases had σ_g values ranging from 2.6 to 3.3, the indicated uniform aerosol size. The low-pressure release σ_g value was higher than all others.

3.3 LOW-PRESSURE DUO RELEASES

A complete listing of the measured values for the uranine mass airborne are tabulated in Table A.5 in Appendix A. Values for the particle size are found in Table A.6 in Appendix A.

3.3.1 Mass Airborne

The average DUO mass airborne is tabulated in Table 3.8. Both the table and figure include average releases of previous higher pressure releases. The low-pressure releases show a greater dependence on pressure on a log-log scale than those at higher pressures. The quantity of powdered uranium airborne falls in the range between the higher pressure releases and previous powder spills.

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TABLE 3.8. Average DUO Mass Airborne from Pressurized Powder Releases

Chamber Pressure, psig	350-g Source DUO, g	100-g Source DUO, g
500.0	30.2	19.8
250.0	20.5	13.0
50.0	6.9	3.6
24.5	0.61	0.30
17.5	0.39	0.06
9.0	0.02	0.03

3.3.2 Weight Percent Airborne

The weight percent airborne values generated in these experiments are listed in Table 3.9 and plotted in Figure 3.4. For all but the 17.5-psig release, the weight percent for the smaller source shows that a larger portion becomes airborne; this is probably a result of the enclosure effect. A larger source requires more distance to disperse. Weight percent of powdered uranium airborne falls in the range between the higher pressure releases and previous powder spills.

3.3.3 Particle Size Distribution and Powder Ejected

Average values of the mass median AED are tabulated in Table 3.10. The release at 24.5 psig gave the highest median aerodynamic equivalent diameter for both source volumes.

At 24.5 psig, 79 to 93% of the DUO powder was ejected. At 17.5 psig, the range dropped to 35 to 86%. At 9 psig, only 24% was ejected.

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TABLE 3.9. Average Weight Percent Airborne from Pressurized DUO Releases

Chamber Pressure, psig	350-g Source	100-g Source
500.0	9.0	20.0
250.0	6.0	13.0
50.0	2.0	4.0
24.5	0.2	0.3
17.5	0.1	0.06
9.0	0.006	0.03

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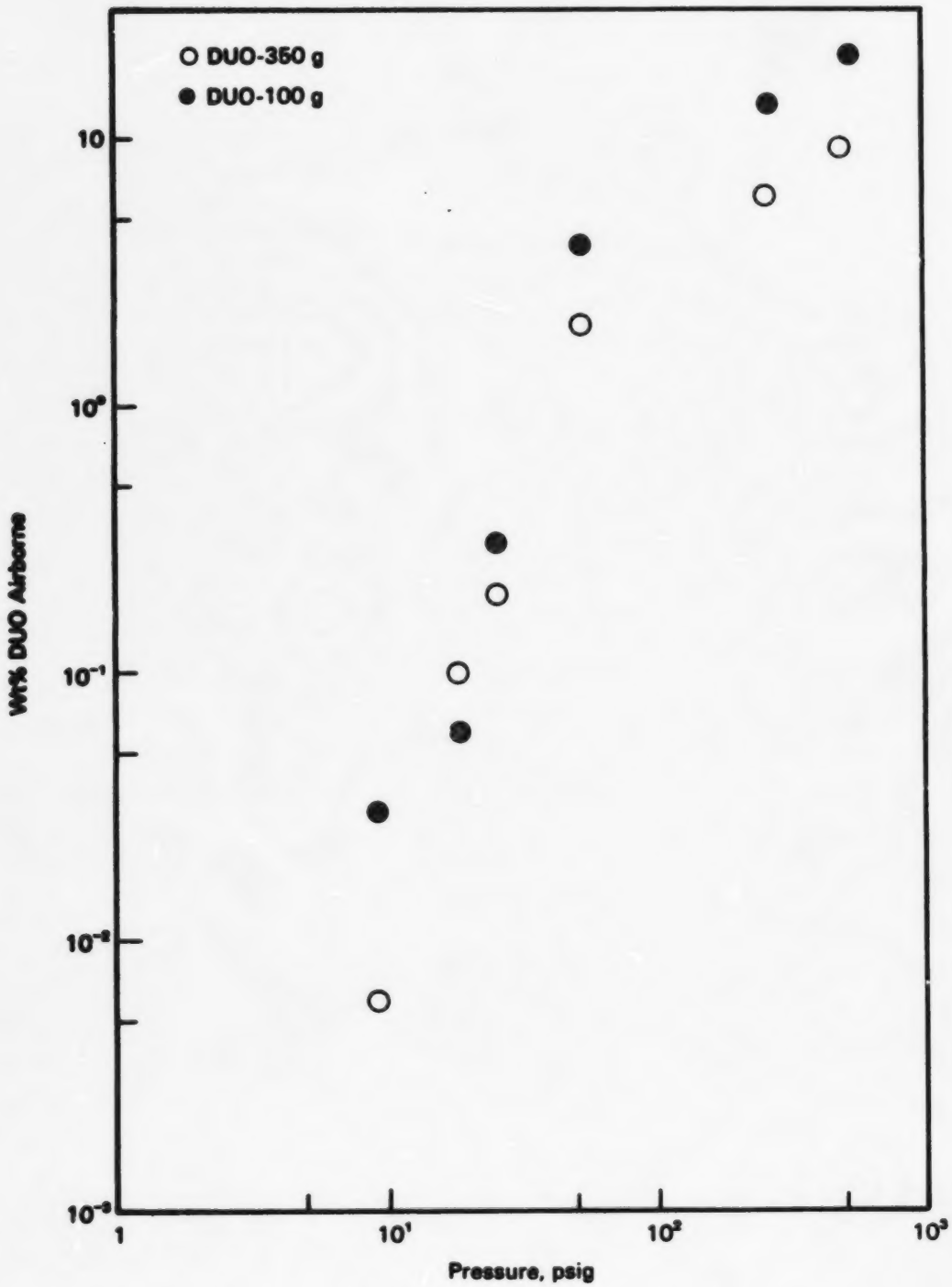

FIGURE 3.4. Weight Percent DUO Airborne Versus Pressure

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TABLE 3.10. Average Median AED of Particles Generated by Pressurized Powder Releases, μm

Chamber Pressure, psig	350-g Source	100-g Source
500.0	16	14
250.0	15	11
50.0	19	19
24.5	38	24
17.5	19	6
9.0	6	7

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APPENDIX A

EXPERIMENTAL DATA

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APPENDIX A

EXPERIMENTAL DATA

TABLE A.1. Mass Airborne from CO_2-Pressurized Releases of Uranine Solutions

Source Volume, cm^3	Pressure, psig	Uranine Collected, g	Wt% Airborne	Total Volume Airborne,[a] cm^3	Volume not Ejected from Chamber, cm^3	Volume Ejected but not Airborne[b]
350	500	4.7×10^{-3}	0.13	0.46	52	297.54
	250	1.4×10^{-3}	0.039	0.14	125	224.74
	50	4.9×10^{-5}	0.0014	0.005	325	25.0
100	500	2.2×10^{-3}	0.22	0.22	48[c]	51.78
	250	5.0×10^{-3}	0.50	0.50	67	32.5
	50	5.5×10^{-5}	0.0055	0.0055	95	5.0

(a) Uranine solution density 0.99 g/cm^3.
(b) Estimated from volume airborne and amount not ejected from PARE.
(c) Under purge ring, modified after run.

TABLE A.2. Median AED of Particles Produced by CO_2-Pressurized Releases of Uranine Solutions

Source Volume, cm^3	Pressure, psig	Mediam Diameter, μm	σ_g	% 10 μm and Less	Wt% of Source that Becomes Airborne and is 10 μm and Less
350	500	3.3	11.0	68	0.088
250	2.3	5.2	82	0.032	
50	1.5	3.8	92	0.0013	
100	500	2.7	5.5	78	0.17
250	2.1	2.6	95	0.48	
50	1.9	3.3	92	0.0046	

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TABLE A.3. Mass Airborne from Flashing-Spray Releases of Superheated Liquids

Source Volume, cc	Pressure, psig	Uranine Collected, cc	Wt% Airborne	Total Volume Airborne, cc	Total Volume not Ejected from Chamber, cc	Volume Ejected but not Airborne, cc
700	125	9.7×10^{-2}	1.51	7.7		692.3
350	240	1.6×10^{-1}	4.96	10.5		339.5
	1257.1×10^{-2}	2.22	17.2		332.8	
	602.8×10^{-2}	8.92	3.1	83	263.9	
100	125	7.8×10^{-2}	8.51	8.5		91.5

(a) Uranine solution density 1.0 g/cc.
(b) Estimated from volume airborne and amount not ejected from PARE.

TABLE A.4. Median AED of Particles Produced by Flashing-Spray Releases of Superheated Uranine Solutions

Source Volume, cc	Pressure, psig	Median Diameter, μm	σ_g	% 10μm and Less	Wt% of Source that Becomes Airborne and is 10 μm and Less
700	125	5.9	2.6	78	1.18
350	240	6.2	2.7	73	3.63
	125	7.7	2.7	66	1.46
	60	8.4	5.6	62	5.53
100	125	6.4	3.3	69	5.87

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TABLE A.5. Mass Airborne from Low-Pressure DUO Releases

Source Mass, g	Pressure, psig	Uranium Collected, g	Wt% Airborne	Total Mass not Ejected from Chamber, g	Mass Ejected but not Airborne, g
350	24.5	0.610	0.174	24.4	325.0
	17.5	0.387	0.111	50.1	299.4
	9.0	0.017	0.005	269.6	80.4
100	24.5	0.302	0.302	21.1	78.7
	17.5	0.63	0.63	65.5	34.4
	9.0	0.03	0.03	75.9	24.1

TABLE A.6. Median AED of Particles Producted by Low-Pressure DUO Releases

Source Mass, g	Pressure, psig	Median Diameter, μm	σ_g	% 10 μm and Less	Wt% of Source that Becomes Airborne and is 10 μm and Less
350	24.5	38	9.3	35	0.061
	17.5	19	6.5	60	0.067
	9.0	6	6.0	61	0.003
100	24.5	24	7.5	42	0.127
	17.5	6	4.6	72	0.045
	9.0	7	4.8	62	0.02

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APPENDIX B

CALCULATIONS

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APPENDIX B

CALCULATIONS

According to Henry's Law (Hougen et al. 1958), the mole fraction of a soluble gas is directly proportional to the partial pressure of that gas above the liquid surface. However, at high pressures (above 1 atmosphere) the value of Henry's Law, constant H, varies with temperature and pressure. In this appendix, the value of H is calculated for the temperatures and pressures in the pressurized release experiments. The amount of gas dissolved in the liquid is calculated for the CO_2- and air-pressurized releases and flashing sprays.

B.1 THE CO_2-H_2O SYSTEM

Pressurized releases using CO_2 as the pressurized gas were run at temperature and pressure ranges of 13 to 21°C and 50 to 500 psig, respectively. Perry's Handbook (Perry et al. 1963) contains data on solubility of CO_2 in H_2O at elevated temperatures and pressures. Lange's Handbook of Chemistry (Lange and Forker 1944) contains additional data on CO_2 solubility in H_2O at 1 atmosphere. Data from Lange is plotted in Figure B.1 and 1/H values taken from the plot at 12, 18, and 25°C to add to data from Perry. Table B.1 lists information from both Lange and Perry.

Mole fraction CO_2 in H_2O is calculated from weight CO_2 per 100 weight H_2O by converting to moles and dividing moles CO_2 by total moles. Calculations of 1/H are made using Henry's Law:

$$N = \left(\frac{1}{H}\right) P$$

where N is the mole fraction of dissolved gas, P is the partial pressure of gas and atmospheres, and H is the mole fraction per atmosphere.

Figure B.2 is a plot of 1/H versus partial pressure of CO_2 at the three temperatures that span experimental temperatures. For experimental pressures of 50, 250, and 500 psig, 1/H for CO_2 is read off the graph at each temperature and listed in Table B.2. The mole fraction of dissolved gas (N) is then calculated for each experimental pressure using Henry's Law. This value is also given in Table B.2. Figure B.3 is a plot of mole fraction of CO_2 in uranine versus temperature at each experimental pressure. From this figure,

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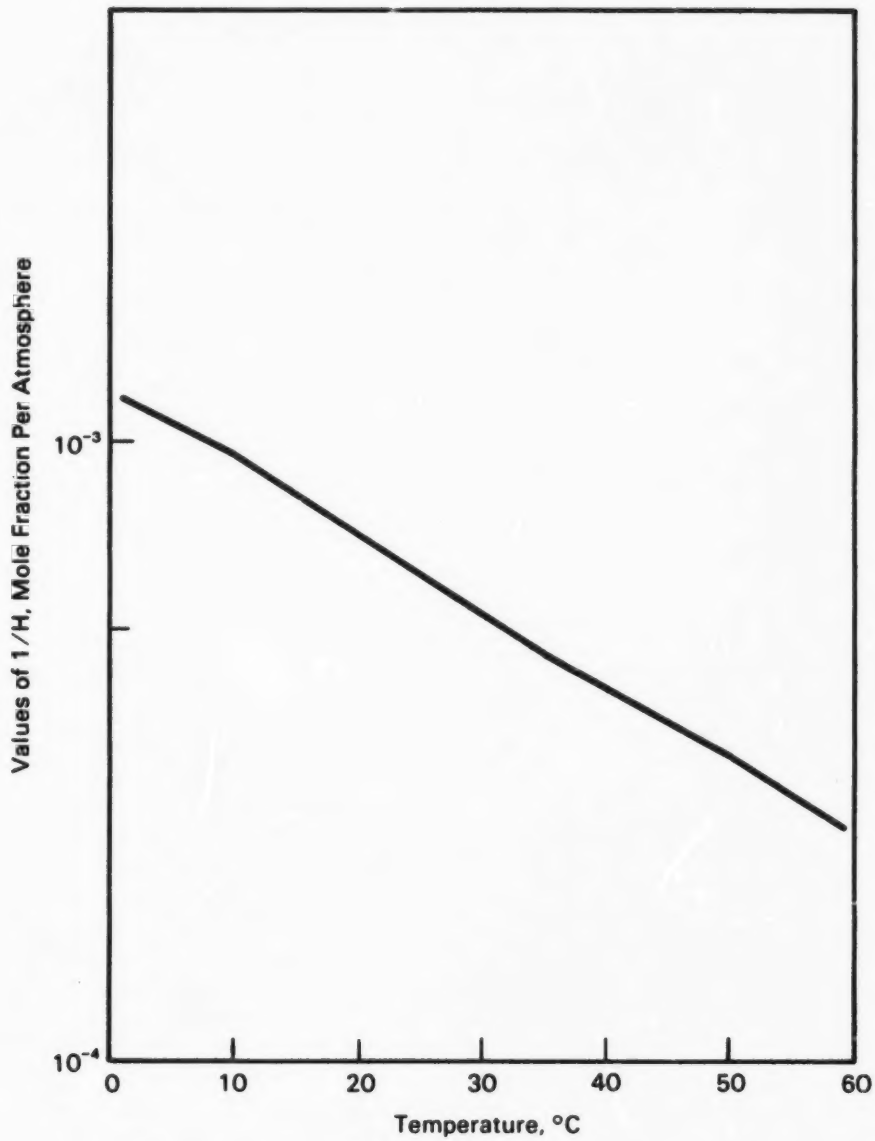

FIGURE B.1. Plot of 1/H Versus Temperature for CO_2 Solubility in H_2O at 1 Atmosphere

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TABLE B.1. Mole Fraction and 1/H by Temperature and Pressure

Total Pressure Atmosphere	Temperature, °C	Wt% CO_2 per 100 Wt% H_2O	Mole Fraction CO_2 in H_2O	1/H, Mole Fraction Per Atmosphere
1	12			8.80×10^{-4}(a)
	18			7.35×10^{-4}(a)
	25			5.95×10^{-4}(a)
25	12	4.5(b)	1.81×10^{-2}	7.23×10^{-4}
	18	3.86	1.55×10^{-2}	6.22×10^{-4}
	25	3.22(b)	1.30×10^{-2}	5.20×10^{-4}
50	12	7.03	2.80×10^{-2}	5.59×10^{-4}
	18	6.33	2.52×10^{-2}	5.05×10^{-4}
	25	5.38	2.15×10^{-2}	4.31×10^{-4}
75	12	7.18	2.85×10^{-2}	3.80×10^{-4}
	18	6.69	2.66×10^{-2}	3.55×10^{-4}
	25	6.17	2.46×10^{-2}	3.28×10^{-4}

(a) Taken directly from Figure B.1.
(b) Interpolated.

the mole fraction of CO_2 dissolved in uranine can be read at the temperature for each experiment; Table B.3 gives these values.

B.2 AIR-H_2O

Perry's Handbook (Perry et al. 1963) provides data on the solubility of nitrogen and oxygen in water. Figure B.4 is a plot of H versus partial pressure for oxygen and nitrogen systems, respectively. The partial pressure of nitrogen and oxygen at experimental pressures is equal to the mole fraction (0.79 and 0.21, respectively) multiplied by the experimental pressure. Table B.4 lists the partial pressures of O_2 and N_2, the value of H from Figure B.4 and the mole fraction of air dissolved in liquid for each experimental pressure.

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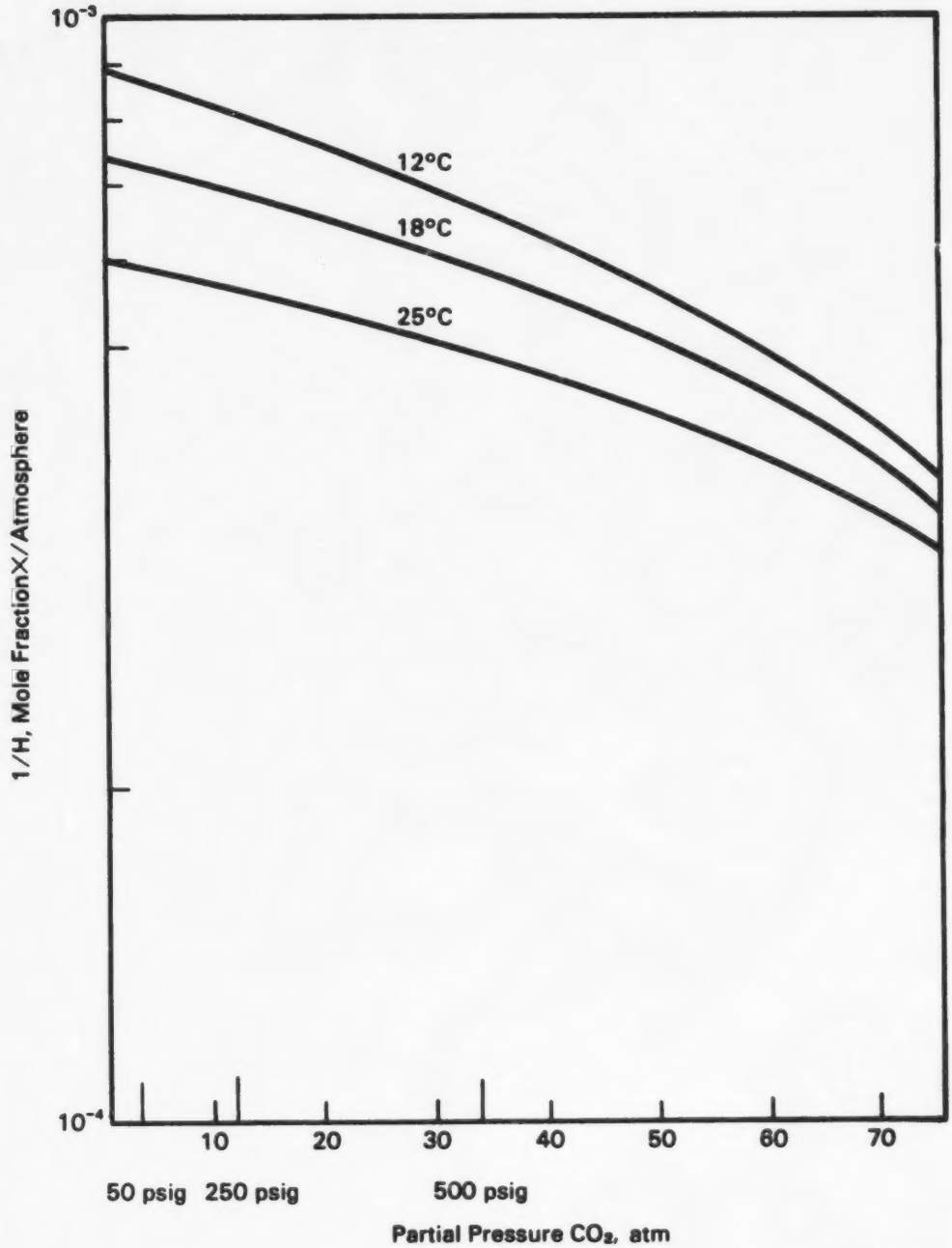

FIGURE B.2. 1/H Versus Partial Pressure of CO_2 for Temperature Range of Experiments

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TABLE B.2. Mole Fraction of CO_2 at Experimental Pressures

CO_2 Partial Pressure Over System		Temperature, °C	1/H Mole Fraction/ Atmosphere	Mole Fraction
psig	atm			
50	3.4	12	8.68×10^{-4}	0.00295
		18	7.25×10^{-4}	0.0025
		25	5.90×10^{-4}	0.0020
250	17	12	8.10×10^{-4}	0.0138
		18	6.88×10^{-4}	0.0117
		25	5.64×10^{-4}	0.0096
500	34	12	6.65×10^{-4}	0.0226
		18	5.88×10^{-4}	0.0200
		25	4.92×10^{-4}	0.0167

B.3 FLASHING SPRAYS

For flashing sprays, the mole fraction of gas dissolved in the liquid is equal to the fraction of vapor flashed at release. The enthalpy and volume of vapor and liquid before and directly at release are used to calculate the fraction flashed. Properties of the liquid in the PARE just before a release of 350 mL of uranine at 163°C are given as the following:

where $T = 163°C = 325°F$
$V_g = 4.66\ ft^3/lbm = 290.7\ mL/g$
$H_g = 1186.2\ Btu/lbm = 2.61\ Btu/g$
$V_L = 0.0177\ ft^3/lbm = 1.10\ mL/g$
$H_L = 294.6\ Btu/lbm = 0.649\ Btu/g$

Total mass in the PARE is 350 g, and PARE volume is 800 mL. If x is the volume of vapor, then 800-x is the volume of liquid and x can be calculated by

$$\frac{x}{290.7} + \frac{800-x}{1.10} = 350$$

$$x = 290.7\ \frac{(350\ (1.1) - 800)}{(1.1 - 290.7)} = 416.6\text{-mL vapor} = 1.43\text{-g vapor}$$

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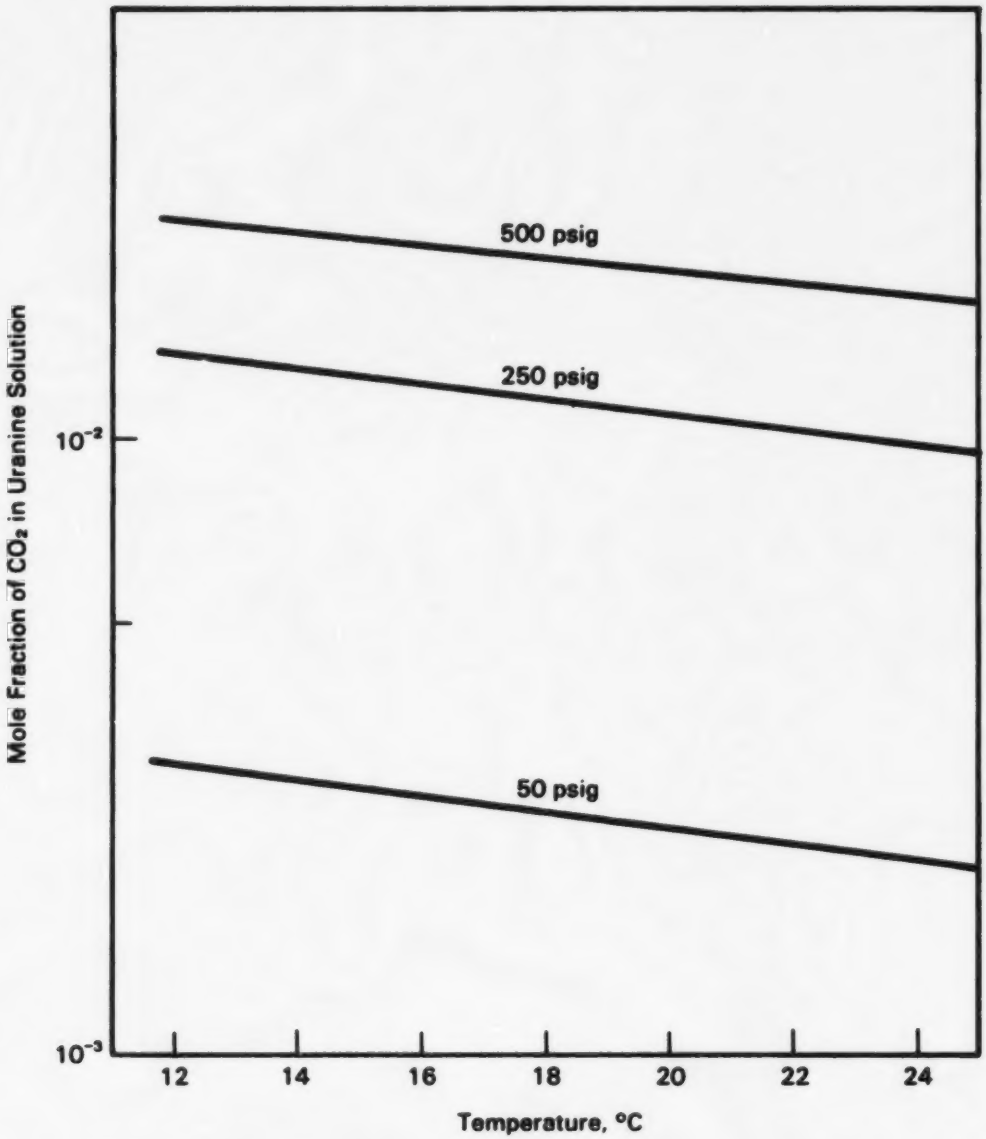

FIGURE B.3. Mole Fraction of CO_2 Dissolved in Uranine Solutions Versus Temperature at Partial Pressure Indicated

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TABLE B.3. Mole Fraction of CO_2 Dissolved in Uranine for CO_2-Pressurized Releases

Run No.	Pressure, psig	Temperature, °C	Mole Fraction	Average Mole Fraction
1	50	21	0.0023	0.0025
6	50	16	0.00265	
3	250	13	0.0135	0.0128
4	250	17	0.0121	
2	500	18	0.0200	0.0200
5	500	18	0.0200	

Liquid volume = 800 - 416.6 = 383.4 mL liquid = 348.5 g liquid

Total energy in the PARE is calculated as follows:

$$E = 1.43 \text{ g } (2.61 \text{ Btu/g}) + 348.5 \text{ g } (0.649 \text{ Btu/g}) = 230 \text{ Btu}$$

At release, some of the uranine flashes immediately dropping the temperature to 100°C. Properties of the liquid at this temperature are listed as follows:

$T = 100°C = 212°F$
$V_g = 26.8 \text{ ft}^3/\text{lbm} = 1672 \text{ mL/g}$
$H_g = 1153.5 \text{ Btu/lbm} = 2.54 \text{ Btu/g}$
$V_L = 0.0167 \text{ ft}^3/\text{lbm} = 1.042 \text{ mL/g}$
$H_L = 180.17 \text{ Btu/lbm} = 0.397 \text{ Btu/g}$

If x is the mass of vapor, the 350-x is the remaining liquid mass and an energy balance can be used to determine x.

$$x(2.54 \text{ Btu/g}) + (350 - x)(0.397 \text{ Btu/g}) = 230 \text{ Btu}$$

$$x = \frac{230 - 350(.397)}{2.54 - 0.397} = 42.5\text{-g vapor}$$

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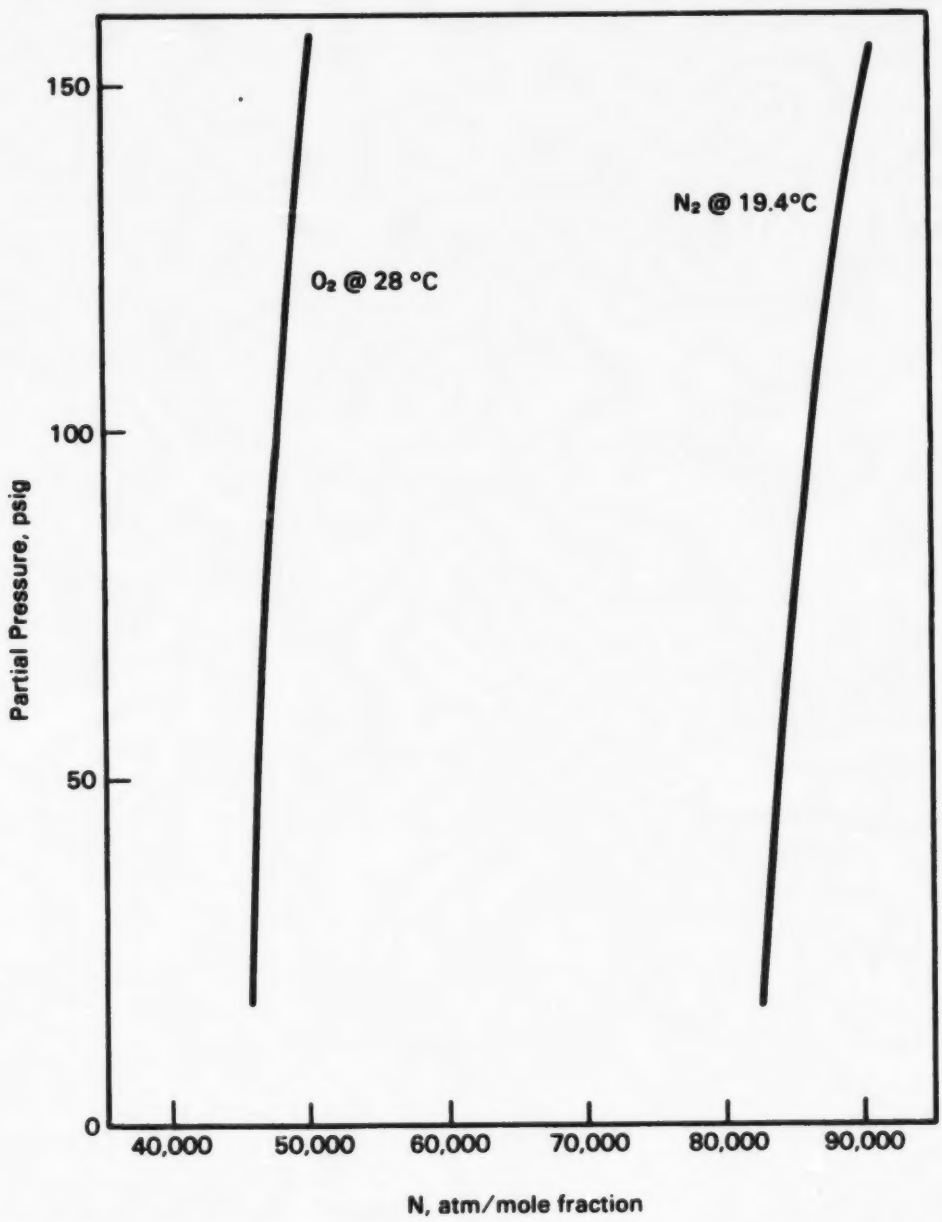

FIGURE B.4. Plot of Henry's Constant, H, Versus Partial Pressure of Oxygen and Nitrogen

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TABLE B.4. Mole Fraction of Air Dissolved in Uranine for Air-Pressurized Releases

Experimental Pressure, psig	Partial Pressure O_2 psig	Partial Pressure N_2 psig	H Atm/Mole Fraction O_2	H Atm/Mole Fraction N_2	Mole Fraction Air in H_2O
50	11	39	45,800	83,200	4.8×10^{-5}
250	53	198	46,000	95,700(a)	2.2×10^{-4}
500	105	395	47,600	112,300(a)	3.9×10^{-4}

(a) Extrapolated values.

Because 1.43 g vapor were present before release, then 42.5 - 1.43 = 41.1 g vapor were flashed during release. The mass fraction of vapor flashed = 41.1 g/348.5 g = 0.12.

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Safety assessments and environmental impact statements for nuclear fuel cycle facilities require an estimate of potential airborne releases. Aerosols generated by accidents are being investigated by Pacific Northwest Laboratory to develop radioactive source-term estimation methods. Experiments measuring the mass airborne and particle size distribution of aerosols produced by pressurized releases were run. Carbon dioxide was used to pressurize uranine solutions to 50, 250, and 500 psig before release. The mass airborne from these experiments was higher than for comparable air-pressurized systems, but not as great as expected based on the amount of gas dissolved in the liquid and the volume of liquid ejected from the release equipment. Flashing sprays of uranine at 60, 125, and 240 psig produced a much larger source term than all other pressurized releases performed under this program. Low-pressure releases of depleted uranium dioxide at 9, 17.5, and 24.5 psig provided data in the energy region between 3-m spills and 50-psig pressurized releases.

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